

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Application No. : 10/594,031 Confirmation No. 4637
Applicant : Kenji SHIGA et al.
5 Filed : September 25, 2006
For : Modifier for Polyester Resin and Process for Producing
Molded Article With the Same
Group Art Unit : 1796
Examiner : Robert Stockton JONES, JR.
10 Docket No. : 12477/13
Customer No. : 23838

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SECOND DECLARATION UNDER 37 C.F.R. 1.132

SIR:

20 I, Kenji Shiga, hereby declare as follows:

1. I received a Bachelor's degree in March 1997 from the College of Chemistry, Ritsumeikan University and a Master's degree in March 1999 from the Graduate School of Science and Engineering, Ritsumeikan University.
- 25 2. I joined the Research Center at Toyobo Co., Ltd. in April 1999. Since then, I have worked in the field of development and research of co-polyester, development and research of polyester resin modifiers, application for adhesives of co-polyesters, and designing molding materials for electronic devices.

3. I am currently employed at Toyobo Co. Ltd. as an assistant manager of the Fundamental Technology & Process Development Department in the Plastics Research and Development Center.

4. I am an inventor for the above-identified patent application; I have read the
5 Office Action mailed July 28, 2010 and the references cited therein; and I am familiar with the subject matter thereof.

5. I have supervised or directed the conduct of the following experiment and hereby submit my report thereon to present further information on the experiment described in the Declaration signed by me on October 21, 2010 (hereinafter referred to
10 as "the first Declaration").

Experiments

1. Composition of Samples (As Described in the First Declaration)

a) Sample A and Example 1 were identical in composition, which composition is shown
15 below:

Amorphous Polyester Resin A (27% by weight) as the first Amorphous Polyester Resin;

Reactive Compound I (3% by weight); and

Amorphous Polyester Resin B (70% by weight) as the second Amorphous Polyester
20 Resin.

b) Sample B and Example 14 were identical in composition, which composition is shown below:

Amorphous Polyester Resin B (27% by weight) as the first Amorphous Polyester Resin;

Reactive Compound J (3% by weight); and

Polyethylene terephthalate (PET) (70% by weight) as the Crystalline Polyester Resin.

c) Sample C and Example 18 were identical in composition, which composition is shown below:

Amorphous Polyester Resin E (20% by weight) as the first Amorphous Polyester Resin;

5 Reactive Compound I (10% by weight); and

Polybutylene naphthalate (PBN) (70% by weight) as the Crystalline Polyester Resin.

The first Amorphous Polyester Resins A, B, and E and Reactive Compounds I and J were prepared as described in the present specification on page 18, line 26 to
10 page 22, line 10.

2. Experimental Procedures

I. The Experiment As Described in the First Declaration

According to pages 2 and 3 of the first Declaration, in Examples 1, 14, and 18,
15 the first Amorphous Polyester Resin A, B, or E and Reactive Compound I or J were first melted and kneaded in order to react only a portion of the two or more glycidyl groups of the reactive compound with the amorphous polyester resin to prepare a modifier for a polyester resin. Then, the modifier and the second Amorphous Polyester Resin B, crystalline polyester PET, or crystalline polyester PBN were melt molded
20 together.

For comparative purposes, Samples A, B, and C were obtained by directly dry-blending

(1) the first Amorphous Polyester Resin A, B, or E,

(2) Reactive Compound I or J, and

25 (3) the second Amorphous Polyester Resin B, crystalline polyester PET, or crystalline polyester PBN to prepare a mixture thereof, without first preparing a modifier for a polyester resin, and then melt molding the mixture.

The conditions for melt molding in Samples A, B, and C corresponded to the conditions for melt molding in Examples 1, 14, and 18, respectively.

II. Additional Experiment

In addition to the experiment described in the first Declaration, the following study and experiment were performed. In the experiment described in the first
5 Declaration,

To experimentally show that only a portion of the glycidyl groups of Reactive Compound I or J (representing reactive compound (II) in the claims) reacted with the first Amorphous Polyester Resin A, first Amorphous Polyester Resin B or first Amorphous Polyester Resin E (representing amorphous polyester resin (I) in the
10 claims), the study and experiment described below were conducted.

As described in Section 2.I. above, in Examples 1, 14 and 18, the first Amorphous Polyester Resin A, B, or E (representing amorphous polyester resin (I) in the claims) and Reactive Compound I or J (representing reactive compound (II) in the claims) were first melted and kneaded to obtain a modifier (V). Then, the modifier (V)
15 and the second Amorphous Polyester Resin B (representing amorphous polyester resin (III) in the claims), crystalline polyester PET, or crystalline polyester PBN (representing crystalline polyester resin (IV) in the claims) were melt molded together.

When amorphous polyester resin (I) having carboxyl groups and reactive compound (II) having glycidyl groups are melted and kneaded together to produce
20 modifier (V), the carboxyl groups react with the glycidyl groups. Because one carboxyl group reacts with one glycidyl group, an equal number of the carboxyl groups and the glycidyl groups are consumed in the reaction.

When x parts by weight of amorphous polyester resin (I) having a carboxyl group concentration CC_I (equivalent/ 10^6 g) and y parts by weight of reactive compound (II) having a glycidyl group concentration CG_{II} (equivalent/ 10^6 g) are melted and
25 kneaded together to obtain $x + y$ parts by weight of modifier (V), the reaction rate of the glycidyl groups on reactive compound (II) is calculated as described below.

If the carboxyl groups of amorphous polyester resin (I) were to NOT react with the glycidyl groups of reactive compound (II), the carboxyl group concentration of

amorphous polyester resin (I) would be simply diluted. Modifier (V) should then have a carboxyl group concentration (equivalent/10⁶g) of $CC_1 \times \{x/(x+y)\}$. In reality, however, a portion of the carboxyl groups of amorphous polyester resin (I) would react with the glycidyl groups of reactive compound (II) when the resin and the reactive compound are melted and kneaded. Accordingly, modifier (V) would have a carboxyl group concentration having a lower value. If the carboxyl group concentration of modifier (V) is measured to be CC_V (equivalent/10⁶g), then $CC_1 \times \{x/(x+y)\} - CC_V$ indicates the amount of carboxyl groups consumed by the reaction. If the carboxyl groups and glycidyl groups do not react, the glycidyl group concentration of modifier (V) is simply diluted, and accordingly, modifier (V) would have a glycidyl group concentration (equivalent/10⁶g) of $CG_{II} \times \{y/(x+y)\}$. Because the concentration of the carboxyl groups consumed by the reaction and that of the glycidyl groups consumed by the reaction are equal, the reaction rate R of the glycidyl groups of reactive compound (II) is represented by:

$$15 \quad R = [CC_1 \times \{x/(x+y)\} - CC_V]/[CG_{II} \times \{y/(x+y)\}].$$

Of the variables used in the above discussion, CC_1 is indicated in Table 1 of the specification (Table 1, the Acid Value row); CG_{II} is indicated as the epoxy values in the synthesis examples of reactive compounds of the examples described in the specification, and x and y are indicated in the specification, tables 2 and 4. Carboxyl group concentration CC_V of modifier (V) is the only variable that is not indicated in the specification. Accordingly, CC_V was obtained through an experiment and the reaction rate of reactive compound (II) was calculated. Note that carboxyl group concentration CC_V of modifier (V) is a value analyzed in ¹H-NMR using nuclear magnetic resonance analyzer "Gemini 200" manufactured by Varian and a solution obtained by mixture of hexafluoroisopropanol-d₂ and chloroform-d.

3. Results

The results are shown in the table below. In Examples 1, 14 and 18, modifier (V) had measured carboxyl group concentration CC_V values of 8 equivalent/10⁶g, 13

equivalent/ 10^6 g and 8 equivalent/ 10^6 g, respectively. These values were used to calculate reaction rates R of reactive compound (II), which were 7%, 7% and 1%, respectively. It is apparent that only a portion of the glycidyl groups of reactive compound (II) was reacted with the carboxyl groups of amorphous polyester resin (I).

Amorphous polyester resin (I)	Reactive compound (II)			Modifier (V)		
	Type	Carboxyl group conc. (eq/10 ³ g)	Amount blended (parts by weight)	Type	Glycidyl group conc. (eq/10 ³ g)	Amount blended (parts by weight)
Ex. 1	A	30	27	1	2627	3
Ex. 14	B	25	27	J	1330	3
Ex. 18	E	20	20	1	2627	10

Carboxyl group conc., as calculated, assuming that no reaction takes place (eq/10³g)

CC_v as measured (eq/10³g)

Carboxyl group conc., as calculated, assuming that no reaction takes place (eq/10³g)

Conc. of Carboxyl groups consumed (eq/10³g)

Glycidyl group conc., as calculated, assuming that no reaction takes place (eq/10³g)

Reaction rate R of glycidyl groups of reactive compound (II) (%)

4. Conclusion

In Examples 1, 14 and 18, it has been clarified that when amorphous polyester resin (I) and reactive compound (II) were melted and kneaded together to obtain a modifier, only a portion of the glycidyl groups of reactive compound (II) was reacted with the carboxyl groups of amorphous polyester resin (I).

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the patent or any reexamination certificate issued therefor.

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Dated: Jan. 17. 2011Kenji Shiga

Kenji Shiga